

# Accelerated leaching of mine spoils as means of reducing specific conductivity

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## Abstract

*Elevated total dissolved solids (TDS) content in waters downstream from surface coal mines in Central Appalachia is an increasing concern, as unnaturally high TDS and/or associated specific conductivity (SC) may contribute to deteriorated stream ecosystem health. The aim of this preliminary study was to explore the efficacy of accelerated leaching as a means of minimizing long-term TDS/SC generation by problematic overburden materials. Five spoil samples were collected and leached using simulated rainwater solutions under ambient and acidified pH conditions. SC and pH were monitored during each leach cycle, and inductively coupled plasma-mass spectroscopy (ICP-MS) was used to determine how leaching conditions affected the release of specific ions. This paper highlights the results of both jar and column leaching tests. Among the most significant findings was that for certain spoil materials, the SC in the leach column effluent was effectively reduced when the spoil was first leached with acidic rainwater followed by ambient rainwater, as opposed to leaching with ambient rainwater alone. This suggests that accelerated leaching might aid in the long-term stabilization of some materials.*

Key words: Specific conductivity, Total dissolved solids, Mine water treatment, Leaching, Mine spoils

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## Introduction

An emerging concern for Central Appalachian streams is the significant increase in total dissolved solids (TDS) and/or associated specific conductivity (SC) occurring downstream of surface mining activities (Hartman et al., 2005; U.S. Environmental Protection Agency, 2011). In particular instances in West Virginia and Kentucky, shifts in the diversity and density of biota downstream of surface coal mines have been correlated with elevated SC relative to reference streams that are not directly affected by mining (Pond et al., 2008; Pond, 2010). Though the exact causal mechanism has been difficult to pinpoint (Fritz et al., 2010; Timpano, 2011), previous studies suggest that unnaturally high TDS and/or SC may contribute to the deteriorated health of such streams (Goodfellow et al., 2000; Weber-Scannell and Duffy, 2007).

Increased TDS downstream of mining activities is fundamentally related to the dissolution of minerals from disturbed geologic materials, such as overburden or other spoils. The relative makeup and concentration of mine-generated dissolved solids, in terms of ions, is therefore dependent on local geology and the degree of interaction between water, as well

as air, and material surfaces. Laboratory-based research indicates that the particular ionic character of a given water likely dictates the major in-stream biologic impacts (Mount et al., 1997; Stekoll, 2003). However, individual ion concentrations as well as TDS are relatively difficult to monitor directly in the field. Instead, SC is commonly used as a surrogate because it is easily measured and is related to TDS (Kennedy et al., 2003). Simply stated, an increased concentration of ions contributed by dissolved solids in turn increases the ability of a water to conduct electricity, though this relationship is not linear because some ions contribute more conductivity than others.

Given that neither SC nor TDS can explicitly define the ionic composition of a water, the use of these parameters to broadly link mining activities with biologic impairments has been criticized (National Mining Association, 2013), as has the development of related regulatory guidance (Cheuse and Chavez, 2013). Nonetheless, on a site-specific basis, changes in TDS and SC in mine water discharge can signal relative changes in the chemical activity, that is, the degree of mineral leaching, of the material influencing that discharge.

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In response to rapidly increasing demands for TDS and/or SC mitigation in Central Appalachia, surface mining and reclamation activities are beginning to account for alterations in these water quality parameters that up to now were not major considerations. Given the rather quick emergence of this issue, the urgent need for results in many cases, and the potential scale of required efforts, innovative solutions are needed.

**Accelerated leaching as an alternative mitigation strategy?** Thus far, strategies to reduce TDS/SC generation or loading to surface waters have focused on materials handling and placement (Taylor et al., 2009), water routing (Palmer and Bernhardt, 2009), or water treatment (*Coal Age*, 2013). Little attention has been paid to the possibility of materials treatment, which could be a viable option. Based on research demonstrating the long-term stabilization of spoil materials by natural weathering processes, and the decline and stabilization of TDS levels in laboratory leach column effluents (Orndorff et al., 2010), an alternative strategy for mitigating long-term TDS generation may be to accelerate the weathering process and stabilize spoil materials over a short period of time.

In one hypothetical situation analogous to metal ore heap leaching, materials could be leached in situ after being placed in a fill (Fig. 1). In this scheme, the dissolved-solids-bearing leach solution, which is the effluent from the fill, could be collected and treated to produce a concentrated brine and a clean leach solution, which could be recycled back through the fill, perhaps with the addition of some fresh reagents or

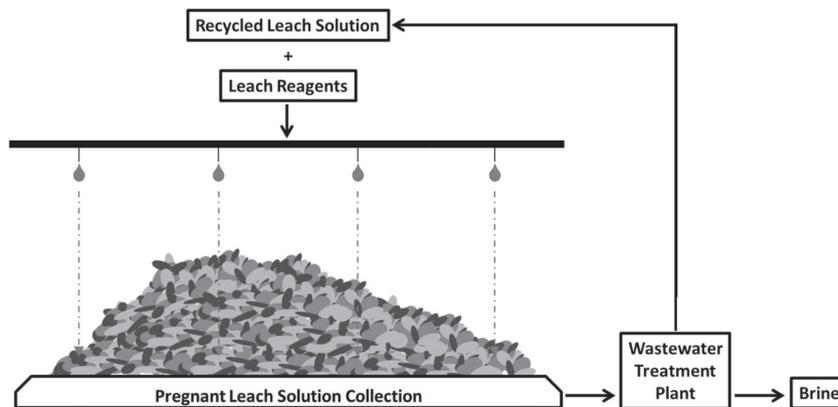
makeup water. The concentrated brine might require further treatment prior to safe disposal or sale.

As a first step in determining the efficacy of an accelerated leaching strategy, this preliminary study investigated the behavior of five typical overburden spoil materials from the Central Appalachian region under several leaching conditions. This paper presents the results of a series of column leach tests, and also summarizes previously unpublished results from jar tests using the same materials.

**Materials and methods**

**Jar tests.** Samples of five spoil materials were collected from surface mines in southern West Virginia, southwestern Virginia and eastern Kentucky. Jar tests were used to create a variety of saturated leaching conditions for Materials A, B, D, E and F, described in Table 1. All test conditions used a ratio of 1 mg material, sized between 250 and 710 µm, to 5 mL leach solution. Fresh materials were initially soaked for four days in ambient simulated rainwater, which was prepared based on work by Halvorson and Gentry (1990) and had the following chemistry: 0.176 mg/L NaCl, 0.709 mg/L CaSO<sub>4</sub>·2H<sub>2</sub>O, 0.355 mg/L MgSO<sub>4</sub>·7H<sub>2</sub>O, 0.069 mg/L K<sub>2</sub>SO<sub>4</sub>, 0.368 mg/L NaNO<sub>3</sub>, 0.746 mg/L (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, 0.243 mg/L NH<sub>4</sub>NO<sub>3</sub>, 0.0084 mg/L H<sub>3</sub>PO<sub>4</sub>. The ambient pH of the simulated rainwater was approximately 5.6.

After the initial soak, the materials were leached for at least five cycles, alternating between conditions designated



**Figure 1** – Proposed leaching and treatment method for removing dissolvable ions from mine spoils.

**Table 1** – Description of initial leach test conditions.

Material	Leaching solution			Description
	Simulated rainwater at ambient pH	Simulated rainwater at pH 2.6	Alternating simulated rainwater at ambient pH/simulated rainwater at pH 2.6	
A	X	X	X	Carbonaceous, no prior weathering.
B	X	X	X	Iron-rich composite sample containing sandstone and black shale. This sample was mildly acidic.
C	X	X		Layered sample consisting of Materials A and B.
D	X	X	X	Composite gray sandstone.
E	X	X	X	Weathered sandstone.
F	X	X	X	Shale.



**Figure 2** – Experimental setup of leaching columns.

as “leaching” and then “resting.” During the leaching cycles, either the ambient simulated rainwater or the simulated rainwater at pH 2.6 was used to fill the jars with the appropriate solution volume, and during the resting cycles, the ambient simulated rainwater was always used. (For Materials A, B and D, several additional leach solutions were initially tested, including some created using deionized water as the base instead of simulated rainwater and some with pH adjusted to 3.6 and 4.6. Based on results from these tests, it was determined that little difference existed between the solutions with a deionized water and simulated rainwater base, or between the ambient simulated rainwater and pH 3.6 or 4.6 conditions. It was therefore decided that all further jar leaching tests would be conducted using simulated rainwater solutions at ambient pH to simulate a real-world environment and an adjusted pH of 2.6 to mimic the most extreme effects of leaching. Only the results obtained using ambient simulated rainwater and simulated rainwater at pH 2.6 are presented here.) The jars were emptied and refilled via a modified “dump and fill” procedure: after dumping the solution from the previous cycle, a small volume of the fresh solution was used to rinse the material by gentle agitation, and then this rinse solution was also dumped and the required volume of fresh solution was added to the jar for the new cycle. Leach cycles lasted two days for Materials A and B and one day for Materials C and D. The pH and SC in each jar were measured daily using an Oakton pH 11 series pH meter and a Hanna HI 9813-6 meter, respectively, while gently stirring the measuring probes just above the spoil materials.

**Initial column leach tests.** A total of 22 leaching columns were constructed using clear acrylic piping with an inner diameter of 2.22 cm. The bottom of each column was covered with a 100-mesh polyester filter to contain the spoil material and then covered with a polyvinyl chloride cap to support the mesh. A 0.25-in. hole was drilled into each of the caps to allow effluent to flow out of the column.

Each material presented in Table 1 was crushed and sized between 0.5 and 3.4 mm and used to fill the columns to a height of 20.32 cm (8 in.). The porosity of each column was determined by pouring water from a graduated cylinder into the column just until the water level reached the top of the spoil material in the column. The amount of water poured from the graduated cylinder was assumed to be equal to the pore space within the column. In order to normalize the leach solution volumes to be used for the tests, experiments were designed to use equal pore volumes of leach solution (rather than equal total volumes) to promote similar saturation of the

material in each column. A peristaltic pump was used to dispense leaching solution onto the top of the material in the columns at a rate of about 10 mL/min. A small plastic funnel and bottle were placed below each column to collect the column effluent. Figure 2 shows the setup of the leaching columns.

During a pretreatment phase, each column was leached with 100 pore volumes of simulated rainwater. The first and last 60 mL of effluent from the columns were collected, and pH and SC were measured. Additionally, metal cations were measured by a Thermo Electron X-Series inductively coupled plasma-mass spectrometry (ICP-MS) system. Following pretreatment, the initial leaching tests were started, during which each column was subjected to regular leach “cycles.” Each cycle involved the application of two pore volumes of a prescribed leach solution (see Table 1): either ambient simulated rainwater or simulated rainwater at pH 2.6. For the “alternating” condition, every other leach cycle was performed with the ambient simulated rainwater, and those in between were performed with simulated rainwater at pH 2.6. To reduce the pH of the simulated rainwater from ambient to 2.6, a 1.375:1 mixture of 96 percent sulfuric acid ( $H_2SO_4$ ) and 70 percent nitric acid ( $HNO_3$ ) was used. Both SC and pH were measured in the effluent from all leaching events, and ICP-MS analysis was conducted during pretreatment and the first four leach cycles, and then intermittently. After the first four ICP-MS samples were prepared, four leach cycles passed before another two ICP-MS samples were made from the next two leach cycles. As testing progressed and the behavior of materials began to stabilize, the frequency at which samples were taken was reduced.

For cycles 0-20 (days 0-35), the columns were leached approximately once per day, and for cycles 21-40 (days 41-50), the columns were leached twice per day, with about eight hours between cycles. (Leaching and measurements were generally only conducted on weekdays, so “daily” leaching refers to that which occurred once per day, Monday through Friday.) Starting with cycle 41 (day 51), all columns were leached only with ambient simulated rainwater. The purpose of this was to determine if the materials originally leached with the pH 2.6 simulated rainwater or alternating solutions had stabilized relative to the materials that had been consistently leached with the ambient simulated rainwater. Between cycles 43 and 44, the columns were allowed to sit and dry for a period of 25 days. After drying, all columns were again leached with simulated rainwater on a daily basis through leach cycle 50 (day 88). Another shorter drying period (10 days) occurred between cycle 50 (day 88) and cycle 51 (day 98). After this point, columns

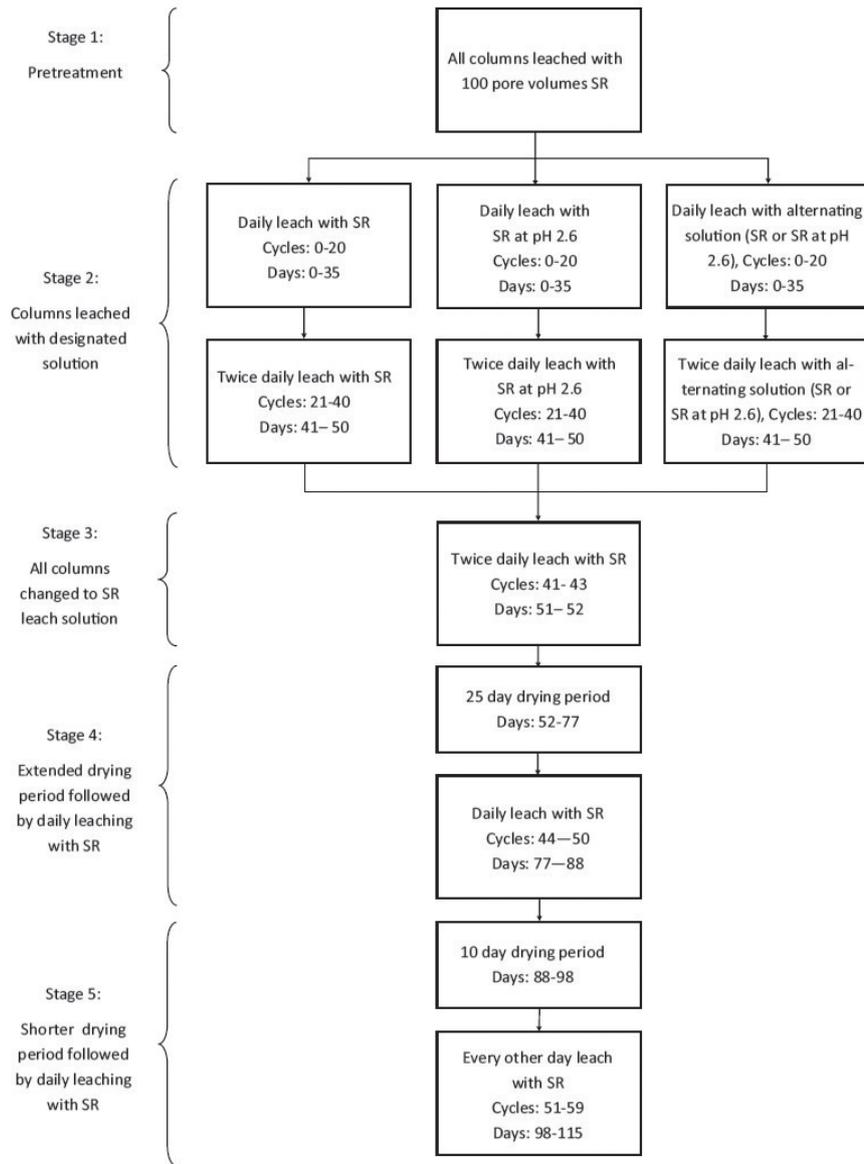


Figure 3 – Flowchart of leach cycles and corresponding leach solutions used.

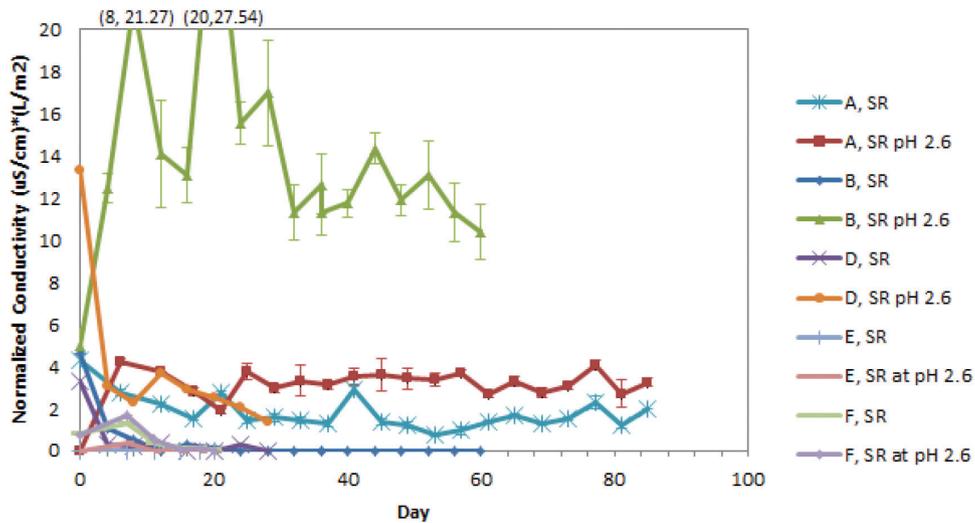


Figure 4 – Normalized SC measured at the end of all resting cycles for jar tests of materials A, B, D, E and F. Points outside of the y-axis range are labeled at the top of the graph (day, normalized conductivity).

were leached every other day with simulated rainwater until the end of the initial leaching experiment at cycle 59 (day 115). Figure 3 shows an overview of the leach cycles and solutions used throughout testing.

**Column leach tests with drying.** After the initial column leaching tests were completed, the columns containing materials A, B, C and D that were leached with simulated rainwater and pH 2.6 simulated rainwater were used to further test the effects of drying the materials between leach cycles. To do this, a new series of leaching and drying cycles were started 53 days after cycle 59 (day 168).

In this series, all columns were leached with two pore volumes of simulated rainwater on a daily basis for 13 days, and pH and SC were measured in the column effluent from each leaching cycle. After leaching, a vacuum pump was used to remove excess water from the columns. A vacuum tube was placed at the bottom of the columns, and each column was vacuumed for two minutes, as no more water could be seen entering the effluent catch-beaker attached to the vacuum after this point. Once all columns had been dewatered, they were placed under an operating fume hood so that air could circulate around the material and promote further drying.

## Results

**Jar tests.** The results of the jar leaching tests are summarized in Fig. 4, which shows the normalized SC in the jars at the end of each resting cycle, just before the jars were dumped and refilled for a leaching cycle. SC was tracked during the resting cycles in order to determine if successive leaching during each leach cycle was indeed progressively stabilizing the materials. Because the size distribution of the different spoil materials, determined using a Microtrac Model X100 particle size analyzer, varied in these tests, some materials had more exposed surface area than others. Thus, SC was normalized by multiplying the measured SC by the ratio of solution volume to material surface area for each jar, giving units of  $\mu\text{S}/\text{cm} \times \text{L}/\text{m}^2$ . Notably, Materials E and F never produced enough TDS for the SC in the resting solution to be measured.

Changes in Material A (carbonaceous) were measured over the longest time period, 20 cycles occurring over 85 days. As these tests were done in triplicate, the error bars in Fig. 4 represent the 95 percent confidence intervals for the mean normalized SC. When leached with simulated rainwater at pH 2.6, the normalized SC in the resting solution

of Material A decreased by one unit to  $3.2 \mu\text{S}/\text{cm} \times \text{L}/\text{m}^2$ , from  $4.2 \mu\text{S}/\text{cm} \times \text{L}/\text{m}^2$ , over the entire test duration. Over the 20 cycles, a slightly larger decrease in SC was seen when ambient simulated rainwater was used as the leach solution, compared with leaching Material A in simulated rainwater at pH 2.6. At the onset of testing, Material A leached in ambient simulated rainwater had an average normalized SC of  $4.3 \mu\text{S}/\text{cm} \times \text{L}/\text{m}^2$ . Leaching over 20 cycles (85 days) decreased the average normalized SC by 2.3 units to  $2.0 \mu\text{S}/\text{cm} \times \text{L}/\text{m}^2$ . For both solutions, the majority of leaching occurred within the first three leach cycles (17 days). In the case of the ambient simulated rainwater leach solution, little change in SC was seen after three cycles. In the case of the simulated rainwater at pH 2.6 leach solution, the SC increased slightly after the third cycle (day 17), then remained fairly constant throughout resting cycles 4 through 20 (days 21-85).

As seen in Fig. 4, exposing Material B (iron-rich composite) to simulated rainwater at pH 2.6 caused an overall increase in the average normalized SC of the resting solution compared with its starting value. Initially, the SC increased sharply from  $5.0$  to  $21.3 \mu\text{S}/\text{cm} \times \text{L}/\text{m}^2$  over the first two leach cycles (8 days). After another SC increase between cycles 4 and 5 (days 16 and 20), the SC gradually decreased for the remainder of the leach cycles. While the SC never returned to its original value, it was still trending downward at the end of the experiment, suggesting that the material may have been slowly stabilizing after an initial activation. This spoil was mildly acidic, and the results indicate that exposing the material to an even more acidic solution enhances the leaching process. Using ambient simulated rainwater as the leach solution quickly resulted in SC being reduced to the point where it could no longer be measured within three leach cycles. It is believed that this condition was “gentle” enough to allow the material to naturally leach, without excess ions being removed as in the pH 2.6 case.

When Material D (gray composite sandstone) was leached in ambient simulated rainwater, a major decrease in SC during the first leach cycle was observed. During this cycle, it appears that nearly all of the dissolvable solids were removed. Before the first leach cycle, Material D in simulated rainwater at pH 2.6 had a normalized specific SC of  $13.3 \mu\text{S}/\text{cm} \times \text{L}/\text{m}^2$ . After the first leach cycle, the SC was reduced to  $3.1 \mu\text{S}/\text{cm} \times \text{L}/\text{m}^2$ . Throughout the remaining leach cycles, the SC slowly decreased to  $1.3 \mu\text{S}/\text{cm} \times \text{L}/\text{m}^2$ , nearly half of the first resting cycle value.

After the first leach cycle, no SC could be measured for Materials E and F.

**Table 2** – Minimum, maximum and average conductivity measurements for samples leached with ambient simulated rainwater.

Material	Measured specific conductivity (ms/cm)				
	Stage 1 (min/max/avg)	Stage 2 (min/max/avg)	Stage 3 (min/max/avg)	Stage 4 (min/max/avg)	Stage 5 (min/max/avg)
A	0.11 ± 0/0.21 ± 0.12/0.16	0.04 ± 0.01/0.23 ± 0.12/0.11	0.10 ± 0.02/0.12 ± 0.02/0.11	0.14 ± 0.01/0.26 ± 0.07/0.19	0.23 ± 0.06/0.29 ± 0.04/0.25
B	0.04/4.13/2.09	0.25/2.51/0.71	0.77/0.85/ 0.82	1.67/5.57/2.73	1.45/3.45/2.02
C	0.02/0.65/0.34	0.02/1.08/0.21	0.13/0.18/0.15	0.19/0.78/ 0.32	0.19/0.40/0.26
D	0.00/0.13/0.07	0.00/0.70/0.08	0.04/0.08/ 0.06	0.09/0.23/0.15	0.16/0.33/0.22
E	0.01/0.19/0.10	0.00/0.00/0.00	0.00/0.00/0.00	0.00/0.01/0.00	0.00/0.00/0.00
F	0.00/0.24/0.12	0.00/0.23/0.04	0.03/0.05/0.04	0.06/0.12/0.08	0.07/0.12/0.09

**Initial column leach tests.** Tables 2-7 detail the SC and major ions present during each stage of leaching shown in Fig. 3. For each leach solution, material and stage, the minimum, maximum and average conductivity values are listed. Because ICP-MS samples were taken less frequently as testing progressed, the minimum, maximum and average ion concentrations for the most prevalent ions are available for stages 1-2 while stages 3-4 only had one ICP-MS sample tak-

en. No ICP-MS samples were taken during stage 5. Detailed results for each material are as follows:

**Material A: Carbonaceous.** The ambient simulated rainwater and pH 2.6 simulated rainwater leaching conditions were run in triplicate. Average SCs for these leach solutions are shown in Fig. 5, and the error bars on the ambient simulated rainwater and pH 2.6 simulated rainwater data points

**Table 3** – Minimum, maximum and average conductivity measurements for samples leached with simulated rainwater at pH 2.6.

Material	Measured specific conductivity (ms/cm)				
	Stage 1 (min/max/avg)	Stage 2 (min/max/avg)	Stage 3 (min/max/avg)	Stage 4 (min/max/avg)	Stage 5 (min/max/avg)
A	0.11 ± 0.01/0.34 ± 0.06/0.22	0.26 ± 0.02/0.42 ± 0.01/0.35	0.15 ± 0.03/0.32 ± 0.12/0.20	0.17 ± 0.03/0.30 ± 0.10/0.22	0.24 ± 0.06/0.30 ± 0.05/0.26
B	0.05/3.32/1.69	0.51/4.29/1.31	1.02/1.39/1.17	1.48/6.66/2.70	1.29/3.47/1.84
C	0.01/ 2.45/1.23	0.09/0.70/0.37	0.13/0.24/0.17	0.23/0.95/0.43	0.23/0.58/0.34
D	0.00/0.30/0.15	0.15/0.38/0.26	0.08/0.26/0.14	0.08/0.21/0.12	0.13/0.26/0.19
E	0.00/0.24/0.12	0.08/0.78/0.51	0.11/0.38/0.22	0.01/0.09/0.04	0.00/0.01/0.00
F	0.00/0.26/0.13	0.15/0.37/0.25	0.06/0.12/0.09	0.03/0.10/0.05	0.03/0.06/0.04

**Table 4** – Minimum, maximum and average conductivity measurements for samples leached with alternating solution. Note that material C was not leached with alternating solution.

Material	Measured specific conductivity (ms/cm)				
	Stage 1 (min/max/avg)	Stage 2 (min/max/avg)	Stage 3 (min/max/avg)	Stage 4 (min/max/avg)	Stage 5 (min/max/avg)
A	0.03/0.37/0.20	0.11/0.40/0.25	0.11/0.15/0.12	0.14/0.24/0.17	0.19/0.26/0.22
B	0.01/2.95/1.48	0.25/1.14/0.50	0.56/0.62/0.59	1.73/4.88/2.61	1.59/ 3.16/2.09
D	0.00/0.08/0.04	0.01/0.33/0.13	0.01/0.03/0.02	0.02/0.07/0.03	0.03/0.06/0.04
E	0.00/0.32/0.11	0.00/0.59/0.20	0.02/0.06/0.04	0.00/0.02/0.00	0.00/0.00/0.00
F	0.00/0.22/ 0.11	0.03/0.26/0.14	0.02/0.00/0.05	0.02 0.06/0.04	0.02/0.05/0.04

**Table 5** – Major ion concentrations present in columns leached with simulated rainwater.

Material	Stage 1 (min/max/avg)	Stage 2 (min/max/avg)	Stage 3 (min/max/avg)	Stage 4 (min/max/avg)
A	<b>Ca:</b> 5.86 ± 1.30/16.63 ± 5.75/11.25 <b>Mg:</b> 210 ± 0.66/7.12 ± 212/4.61 <b>Fe:</b> 4.77 ± 3.58/8.55 ± 14.17/6.66	<b>Ca:</b> 5.06 ± 5.71/24.72 ± 9.63/13.97 <b>Mg:</b> 1.49 ± 0.27/12.20 ± 0.00/5.56 <b>Fe:</b> 0.25 ± 0.07/3.20 ± 4.70/1.05	<b>Ca:</b> 16.61 ± 4.32 <b>Mg:</b> 4.75 ± 1.01 <b>Fe:</b> 0.67 ± 0.48	<b>Ca:</b> 24.42 ± 8.17 <b>Mg:</b> 9.60 ± 1.77 <b>Fe:</b> 1.89 ± 3.17
B	<b>Ca:</b> 3.70/336.50/170.10 <b>Mg:</b> 1.54/279.60/140.57 <b>Fe:</b> 1.72/158.10/79.91	<b>Ca:</b> 1.97/194.90/38.65 <b>Mg:</b> 1.22/122.60/24.95 <b>Fe:</b> 0.00/96.29/22.70	<b>Ca:</b> 9.87 <b>Mg:</b> 8.23 <b>Fe:</b> 11.04	<b>Ca:</b> 126.30 <b>Mg:</b> 133.50 <b>Fe:</b> 1006.00
C	<b>Ca:</b> 58.66/138.80/98.73 <b>Mg:</b> 23.41/38.77/31.09 <b>Fe:</b> 1.08/7.97/4.52	<b>Ca:</b> 5.93/69.21/30.30 <b>Mg:</b> 1.48/24.83/9.44 <b>Fe:</b> 0.14/1.70/0.92	<b>Ca:</b> 20.02 <b>Mg:</b> 4.90 <b>Fe:</b> 0.46	<b>Ca:</b> 94.02 <b>Mg:</b> 23.70 <b>Fe:</b> 0.28
D	<b>Ca:</b> 5.37/33.46/19.42 <b>Mg:</b> 0.90/5.96/3.43 <b>K:</b> 0.07/17.24/8.66	<b>Ca:</b> 2.01/54.74/12.53 <b>Mg:</b> 0.44/24.29/4.00 <b>K:</b> 0.00/3.13/1.13	<b>Ca:</b> 10.25 <b>Mg:</b> 1.92 <b>K:</b> 0.65	<b>Ca:</b> 15.70 <b>Mg:</b> 3.45 <b>K:</b> 1.04
E	<b>Ca:</b> 2.52/3.98/3.25 <b>Mg:</b> 0.77/2.07/1.42 <b>Fe:</b> 1.05/1.48/ 1.26	<b>Ca:</b> 0.51/2.63/1.74 <b>Mg:</b> 0.21/1.27/0.64 <b>Fe:</b> 0.19/3.32/0.70	<b>Ca:</b> 0.97 <b>Mg:</b> 0.40 <b>Fe:</b> 0.21	<b>Ca:</b> 1.90 <b>Mg:</b> 0.79 <b>Fe:</b> 0.63
F	<b>Ca:</b> 2.57/22.02/12.29 <b>Mg:</b> 1.15/12.58/6.87 <b>K:</b> 0.38/6.95/3.67	<b>Ca:</b> 1.93/18.72/6.72 <b>Mg:</b> 0.84/12.10/3.90 <b>K:</b> 0.31/3.99/1.53	<b>Ca:</b> 4.09 <b>Mg:</b> 2.67 <b>K:</b> 1.47	<b>Ca:</b> 9.97 <b>Mg:</b> 6.47 <b>K:</b> 1.83

represent the 95 percent confidence interval for the average of these columns. The effluent SC for the alternating leach condition is also shown in Fig. 5. Of the three initial leaching conditions, the ambient simulated rainwater was the least active. The SC of the alternating condition effluent was consistently between the ambient simulated rainwater and pH 2.6 simulated rainwater conditions. Prior to the 25-day drying period, there was a very slight decrease in the SC of all columns with this material. However, the magnitude of decrease was greater in the columns leached with simulated rainwater at pH 2.6 and alternating leach solutions.

During cycles 41 and 42 (day 51), the columns originally

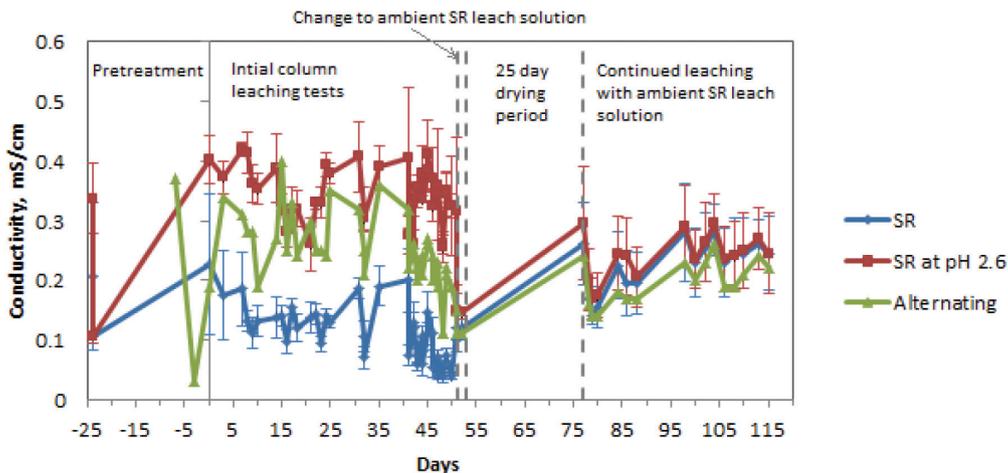
leached with simulated rainwater at pH 2.6 and alternating solutions were changed to the ambient simulated rainwater leach solution. During these two cycles, the SC values of these two columns decreased and were nearly equivalent to the SC of the column traditionally leached with ambient simulated rainwater. After the 25-day drying period, the SC of all three conditions increased to a near equal level. As the columns continued to be leached with ambient simulated rainwater, SC decreased for all columns until cycle 48 (day 84). The 48th cycle follows another drying period spanning the previous weekend, over which leaching did not occur and the materials were allowed to dry and “reactivate.” With consis-

**Table 6** – Major ion concentrations present in columns leached with simulated rainwater at pH 2.6.

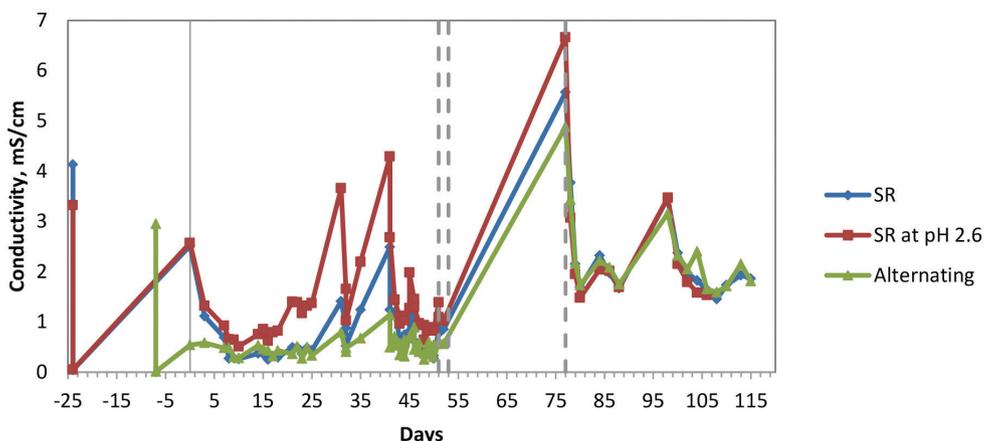
Material	Stage 1 (min/max/avg)	Stage 2 (min/max/avg)	Stage 3 (min/max/avg)	Stage 4 (min/max/avg)
A	<b>Ca:</b> 6.22 ± 0.41/21.90 ± 4.81/ 14.06	<b>Ca:</b> 3.15 ± 2.20/53.07 ± 4.27/34.38	<b>Ca:</b> 38.24 ± 24.86	<b>Ca:</b> 28.25 ± 10.62
	<b>Mg:</b> 2.24 ± 0.16/9.87 ± 2.18/6.05	<b>Mg:</b> 0.97 ± 0.63/23.79 ± 1.87/12.74	<b>Mg:</b> 9.21 ± 5.98	<b>Mg:</b> 9.72 ± 2.33
	<b>K:</b> 1.11 ± 0.36/14.2 ± 2.96/7.61	<b>K:</b> 0.23 ± 0.11/5.30 ± 0.46/2.06	<b>K:</b> 1.58 ± 0.45	<b>K:</b> 1.59 ± 0.25
	<b>Fe:</b> 3.12 ± 3.05/3.42 ± 0.57/ 3.27	<b>Fe:</b> 0.16 ± 0.06/11.58 ± 16.46/2.03	<b>Fe:</b> 0.51 ± 0.41	<b>Fe:</b> 2.56 ± 2.37
B	<b>Ca:</b> 4.78/285.40/145.09	<b>Ca:</b> 0.38/160.5/34.01	<b>Ca:</b> 5.89	<b>Ca:</b> 141.10
	<b>Mg:</b> 1.77/162.67/81.34	<b>Mg:</b> 0.01/ 115.20/23.26	<b>Mg:</b> 4.34	<b>Mg:</b> 139.5
	<b>Fe:</b> 1.55/80.68/41.12	<b>Fe:</b> 0.13/129.30/34.62	<b>Fe:</b> 18.88	<b>Fe:</b> 1385.00
C	<b>Ca:</b> 4.81/233.70/119.26	<b>Ca:</b> 30.54/64.60/44.81	<b>Ca:</b> 15.20	<b>Ca:</b> 105.20
	<b>Mg:</b> 1.91/86.38/44.14	<b>Mg:</b> 8.34/27.80/15.55	<b>Mg:</b> 4.49	<b>Mg:</b> 35.23
	<b>Fe:</b> 5.42/142.50/73.96	<b>Fe:</b> 0.35/8.31/2.18	<b>Fe:</b> 0.38	<b>Fe:</b> 0.30
D	<b>Ca:</b> 5.27/32.12/18.70	<b>Ca:</b> 7.72/48.07/30.87	<b>Ca:</b> 13.33	<b>Ca:</b> 25.61
	<b>Mg:</b> 1.21/5.49/3.35	<b>Mg:</b> 2.80/13.59/6.87	<b>Mg:</b> 2.41	<b>Mg:</b> 4.66
	<b>K:</b> 0.49/16.70/8.59	<b>K:</b> 0.04/3.05/1.31	<b>K:</b> 0.02	<b>K:</b> 0.68
E	<b>Ca:</b> 13.31/23.48/18.40	<b>Ca:</b> 0.35/30.35/11.84	<b>Ca:</b> 0.22	<b>Ca:</b> 0.23
	<b>Mg:</b> 5.61/10.32/7.96	<b>Mg:</b> 0.19/10.47/4.15	<b>Mg:</b> 0.22	<b>Mg:</b> 0.70
	<b>Fe:</b> 0.86 /0.91/ 0.88	<b>Fe:</b> 0.32/4.16/0.99	<b>Fe:</b> 0.26	<b>Fe:</b> 0.19
F	<b>Ca:</b> 3.61 /22.53/13.07	<b>Ca:</b> 16.74/29.92/23.34	<b>Ca:</b> 6.51	<b>Ca:</b> 7.88
	<b>Mg:</b> 1.17/12.76/6.96	<b>Mg:</b> 6.97/19.42/ 13.67	<b>Mg:</b> 4.14	<b>Mg:</b> 5.65
	<b>K:</b> 0.53/6.89/3.71	<b>K:</b> 1.58/6.47/3.19	<b>K:</b> 1.80	<b>K:</b> 1.91

**Table 7** – Major ion concentrations present in columns leached with alternating solutions. Note that Material C was not leached with alternating solution.

Material	Stage 1 (min/max/avg)	Stage 2 (min/max/avg)	Stage 3 (min/max/avg)	Stage 4 (min/max/avg)
A	<b>Ca:</b> 6.50/22.28/14.39	<b>Ca:</b> 21.36/36.63/24.74	<b>Ca:</b> 24.83	<b>Ca:</b> 30.67
	<b>Mg:</b> 2.10/9.93 /6.02	<b>Mg:</b> 1.85/14.33/9.15	<b>Mg:</b> 5.10	<b>Mg:</b> 6.25
	<b>K:</b> 0.34/ 13.23/6.79	<b>K:</b> 0.21/2.67/1.74	<b>K:</b> 0.91	<b>K:</b> 0.95
	<b>Fe:</b> 1.01/3.05/2.09	<b>Fe:</b> 0.24/3.39/0.93	<b>Fe:</b> 0.28	<b>Fe:</b> 0.21
B	<b>Ca:</b> 2.78/252.50/127.64	<b>Ca:</b> 1.73/36.31/18.32	<b>Ca:</b> 10.74	<b>Ca:</b> 116.70
	<b>Mg:</b> 1.22/150.90/76.06	<b>Mg:</b> 0.04/21.51/11.89	<b>Mg:</b> 7.72	<b>Mg:</b> 125.30
	<b>Fe:</b> 0.62/88.50/44.56	<b>Fe:</b> 0.01/8.73/3.78	<b>Fe:</b> 3.01	<b>Fe:</b> 780.50
D	<b>Ca:</b> 4.05/12.94/8.49	<b>Ca:</b> 5.52/54.83/24.06	<b>Ca:</b> 3.35	<b>Ca:</b> 9.05
	<b>Mg:</b> 0.98/2.91/1.94	<b>Mg:</b> 1.17/ 13.45/5.22	<b>Mg:</b> 0.76	<b>Mg:</b> 2.08
	<b>K:</b> 1.06/6.12/3.59	<b>K:</b> 0.00/2.21/0.75	<b>K:</b> 0.17	<b>K:</b> 0.51
E	<b>Ca:</b> 3.22/30.91/17.07	<b>Ca:</b> 1.03/17.64/6.79	<b>Ca:</b> 0.36	<b>Ca:</b> 0.62
	<b>Mg:</b> 1.50/14.26/7.88	<b>Mg:</b> 0.29/6.74/2.22	<b>Mg:</b> 0.14	<b>Mg:</b> 0.28
	<b>Fe:</b> 0.83/1.70/1.26	<b>Fe:</b> 0.22/1.84/0.63	<b>Fe:</b> 0.22	<b>Fe:</b> 0.38
F	<b>Ca:</b> 3.97/18.44/11.21	<b>Ca:</b> 7.36/ 27.49/14.11	<b>Ca:</b> 1.3	<b>Ca:</b> 5.10
	<b>Mg:</b> 1.67/11.16/6.42	<b>Mg:</b> 4.36/19.13/9.22	<b>Mg:</b> 2.43	<b>Mg:</b> 3.26
	<b>K:</b> 1.05/6.75/3.90	<b>K:</b> 0.34/4.12/2.22	<b>K:</b> 1.91	<b>K:</b> 0.98



**Figure 5** – Specific conductivity of leach effluents from columns containing Material A. Significant changes in experimental conditions are noted on the graph for reference.



**Figure 6** – Specific conductivity of leach effluents from columns containing Material B.

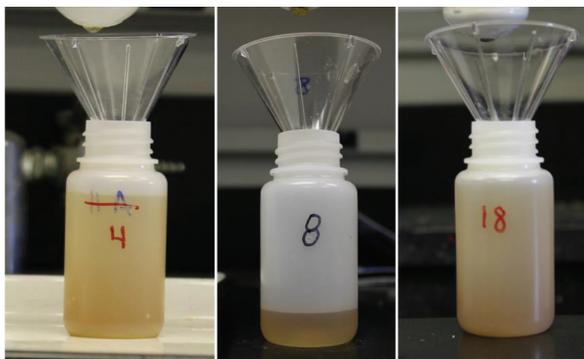
tent leaching following the weekend, SC values continued to decrease until a longer, 10-day drying period between cycles 50-51 (days 88-98). After the 10-day drying period, the SC stayed relatively constant for the three columns.

**Material B: Iron-rich composite.** Unlike Material A, the column with Material B, the acidic and iron-rich composite, leached with alternating solutions (Fig. 6), behaved

quite similarly to the column with Material B leached with the ambient simulated rainwater alone. Alternating the leach solution between ambient simulated rainwater and pH 2.6 simulated rainwater caused the SC in the effluent to be fairly constant throughout the initial testing, prior to drying. Leaching back-to-back with ambient simulated rainwater between cycles 40 and 42 (days 50 and 51) had little measurable effect on the effluent’s SC.

Over time, the column leached with simulated rainwater at pH 2.6 had higher SC than the column leached with ambient simulated rainwater, but they exhibited similar trends. During the initial leach cycles, there were decreases in SC for both effluents. The SC of the effluent of the ambient simulated rainwater column remained constant until cycle 15. Between cycles 15 and 32 (days 24-46), the SC of the effluent was slightly elevated compared with the initial values between cycles 5 and 15 (days 9-24). This trend is shown by the blue line (diamond marker) in Fig. 6. Similar trends were seen in the simulated rainwater at pH 2.6 column, but the increase in SC was seen between cycles 5 and 33 (days 9-47).

All three columns had a large increase in SC (and a sharp decrease in pH) after the 25-day drying period. Increases of 4.72, 5.64 and 4.32 mS/cm were seen in the columns for ambient simulated rainwater, pH 2.6 simulated rainwater and alternating solutions, respectively. The increased SC for all of the columns was quickly tempered with each successive



**Figure 7** – Leach effluents collected from the iron-rich composite sample after a 25-day drying period. ICP-MS analysis showed high concentrations of iron.

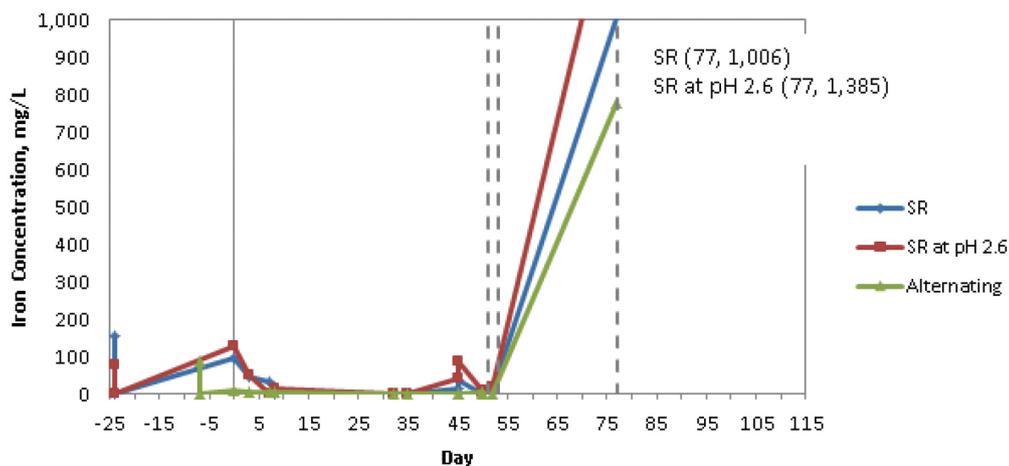
leaching with simulated rainwater. After a weekend period marked by cycle 48 (day 84), the columns had reactivated slightly, causing an increase in SC. Again, this trend was short-lived, and the SC values then decreased with the every-other-day leaching schedule, though they never returned to the lower values measured prior to the 25-day drying period.

Once the leach effluents were collected from the columns, there was a dramatic difference in the color of the effluents. As suggested by the reddish color of the leach effluent shown in Fig. 7, the samples had extremely high iron contents after the drying period. For comparison, Fig. 8 shows the change in iron concentration after the 25-day drying period. After the drying period, the iron content was highest in the sample leached with simulated rainwater at pH 2.6, measuring 1,385 mg/L, followed by the ambient simulated rainwater and alternating solutions at 1,006 and 780 mg/L, respectively. While iron was by far the most abundant element present after the drying period, increases in aluminum, magnesium and calcium were also present.

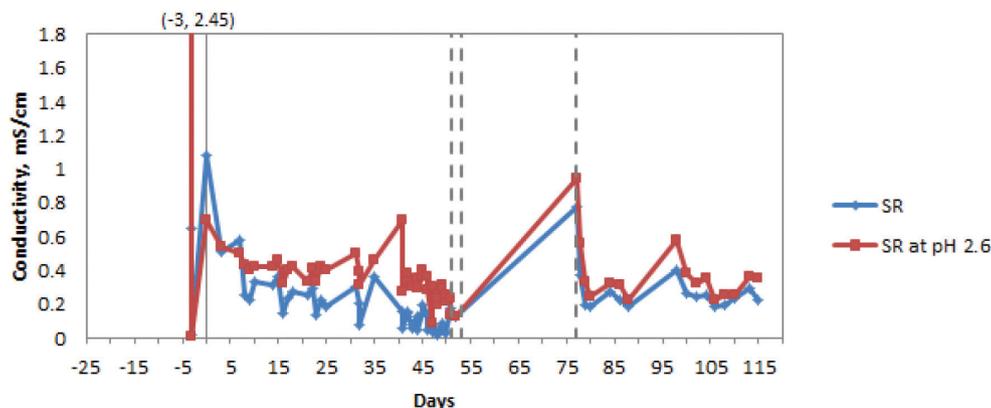
**Material C: Layered iron-rich composite and carbonaceous.** Columns were also constructed in which Materials A and B were layered to determine overall effects on SC. Two layers of each material were used, in the order B, A, B, A from the bottom of the column to the top, and each layer was approximately 4.5 cm thick. In these columns, only the ambi-

ent simulated rainwater and pH 2.6 simulated rainwater conditions were tested. Though the ambient simulated rainwater condition resulted in slightly lower SC than the simulated rainwater at pH 2.6, Fig. 9 shows that the two columns behaved very similarly. After the 25-day drying period marked by cycle 44 (day 77), spikes can be seen in the SC of both effluents, but they are of much smaller magnitude than those seen for Material B (Fig. 6). After continued leaching of the columns, the SC measurements returned to the same level as before the 25-day drying period. This result was not seen in any of the columns filled with only Material A or Material B.

The SC measurements from the columns containing Material C fell between those containing Materials A and B, but tended to be nearer to the measurements taken from the columns containing Material A, as seen in Figs. 10 and 11. Similar to Material A alone, the layered columns had the highest concentrations of calcium, magnesium and potassium throughout leaching. The calcium concentrations, however, in the Material C columns remained between those measured in the columns containing Materials A and B. Like Material B alone, spikes in the concentrations of calcium and magnesium were seen after the drying period, although increases in SC for Material C were of much smaller magnitude than the increases observed for the Material B columns. Interestingly, the increase in iron concentrations that was observed for Material B after drying was not seen for Material C.



**Figure 8** – Change in iron concentration for iron-rich composite sample after the 25-day drying period. Concentrations on day 77 were far outside of the y-axis range and are noted on the right of the graph.



**Figure 9** – Specific conductivity of leach effluents from columns containing Material C (layered carbonaceous and iron-rich composite materials). Points outside of the y-axis range are labeled at the top of the graph.

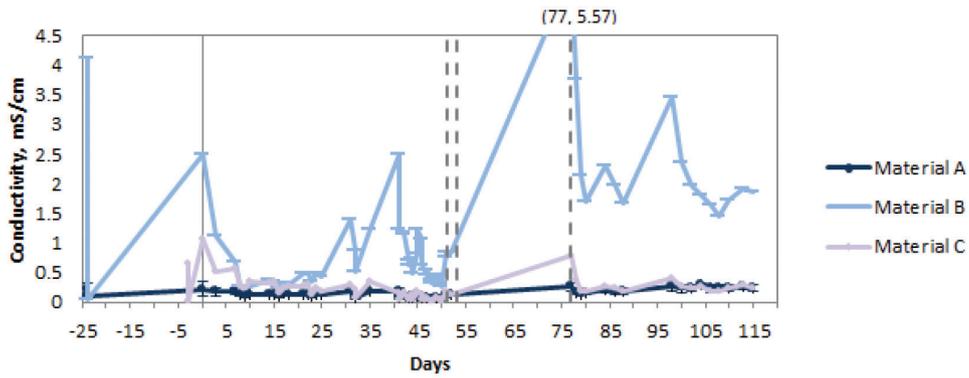


Figure 10 – Comparison of SC for Materials A, B and C when ambient simulated rainwater was used as the leach solution. Points outside of the y-axis range are labeled at the top of the graph.

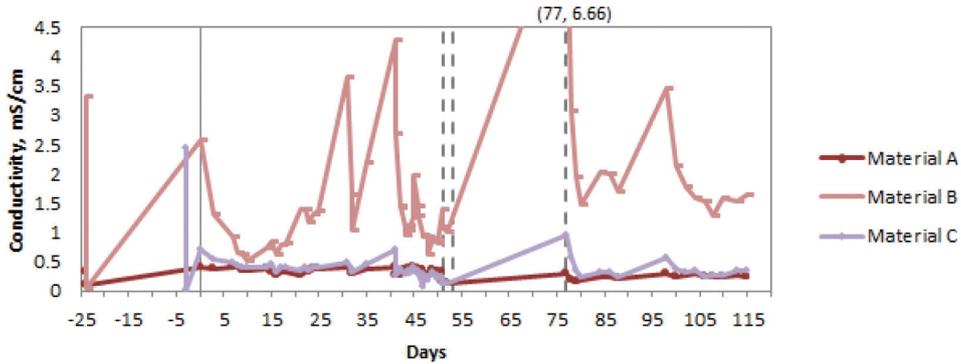


Figure 11 – Comparison of SC for Materials A, B and C when simulated rainwater at pH 2.6 was used as the leach solution. Points outside of the y-axis range are labeled at the top of the graph.

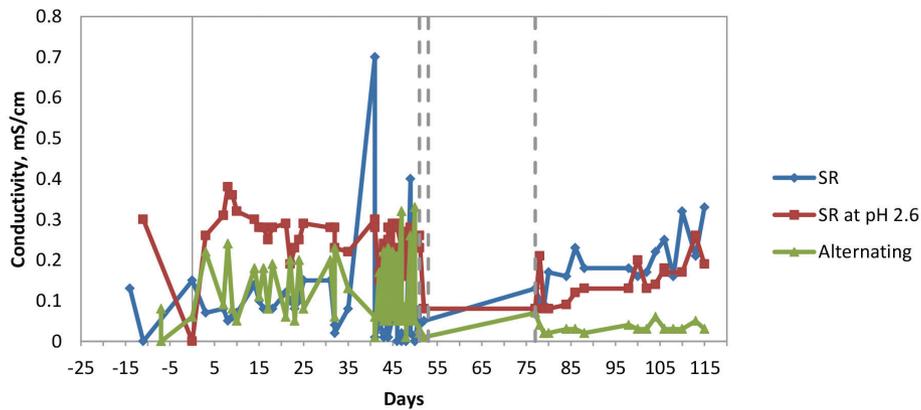


Figure 12 – Specific conductivity of leach effluents from columns containing Material D.

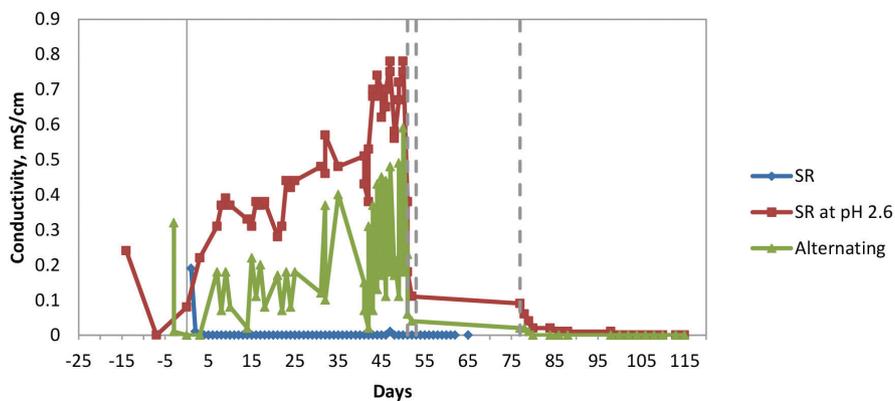


Figure 13 – Specific conductivity of leach effluents from columns containing Material E.

**Material D: Composite gray sandstone.** For Material D (composite sandstone), the SC measurements of the alternating leach solution effluent were highly dependent on the type of leach solution that was being used. This trend is shown by the zig-zag shape of the SC measurements in Fig. 12. When the alternating solution column was leached with simulated rainwater at pH 2.6, the SC values were nearer to those of the column always leached with simulated rainwater at pH 2.6 solution. During the next leach cycle with ambient simulated rainwater, the SC values were close to the SC values of the column consistently leached with ambient simulated rainwater. The column leached with alternating solution showed little change in SC until the solution was changed to ambient simulated rainwater on day 51 (cycle 41), after which the SC decreased to nearly 0 mS/cm.

The 25-day drying period between days 52 and 77 caused only a slight increase in the SC of the columns compared with the SC value just before drying. However, the SC in the ambient simulated rainwater and pH 2.6 simulated rainwater columns continued to rise during the leach cycles after the dry-out. Exciting results were viewed in the column originally leached with the alternating solutions. After the change to the ambient simulated rainwater only solution, the alternating solutions column had consistently low SC, averaging 0.03 mS/cm. It is therefore believed that the alternating solutions may have helped accelerate the leaching of soluble ions within Material D. Additionally, it can be seen that the ambient simulated rainwater column had higher SC measurements after drying compared with the simulated rainwater at pH 2.6 and alternating solutions columns

**Material E: Weathered sandstone.** As seen in Fig. 13, after pretreatment, SC measurements indicated that little if any leaching of Material E (weathered sandstone) occurred with the ambient simulated rainwater treatment. The columns leached with the simulated rainwater at pH 2.6 and alternating solutions became more active until the leach solution was changed to ambient simulated rainwater in cycle 41 (day 51). While both had increasing activity over time, the column leached with simulated rainwater at pH 2.6 was more active than the column leached with the alternating solution. As with Material D (composite gray sandstone), the SC of the alternating condition effluent was highly dependent on which solution was used during the leach cycle. Higher conductivities occurred when simulated rainwater at pH 2.6 solution was used, while lower conductivities occurred when ambient simulated rainwater was used. As seen in Fig. 13, once all columns were switched to ambient simulated rainwater

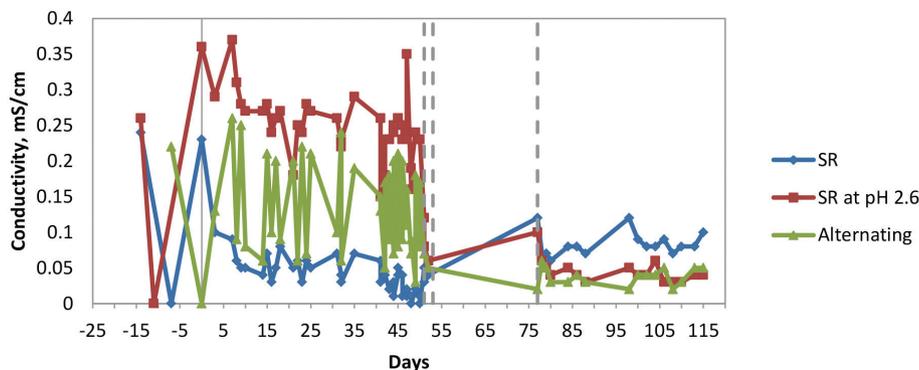
during cycle 41 (day 51), the SC of the effluents quickly decreased. None of the columns showed an increase in SC due to the 25-day drying period.

**Material F: Shale.** Over time, all of the columns containing Material F (shale) showed a decreasing trend in SC, regardless of the leach solution used (Fig. 14). As with the sandstone samples (Materials D and E), the SC of the effluent from the column leached with the alternating solution was dependent on the solution used during leaching. Once the columns were leached with ambient simulated rainwater only, the SC of the columns for the pH 2.6 simulated rainwater and alternating solutions began to decrease while the column traditionally leached with ambient simulated rainwater started to have increasing SC.

During cycle 45 (day 78), the effluent SC of the columns for the pH 2.6 simulated rainwater and alternating solutions fell below levels measured in the ambient simulated rainwater column. It is believed that leaching these samples with the acidic solutions accelerated the release of ions that are responsible for increasing SC values.

**Column leach tests with drying.** Results for the column leach tests with drying are shown in Figs. 15-18. For reference, the graphs show the 14 leach cycles leading up the end of the initial column leaching tests along with the 14 cycles during which dewatering was performed.

No major increases in SC between leach cycles were observed. However, for all of the materials, there was a large increase in SC between the end of the initial column leaching tests and the start of the column leaching tests with drying. The largest increase in SC between the two sets of tests was observed for Material B, the iron-rich composite (Fig. 16). During the last cycle of the initial column leaching tests, the column leached with ambient simulated rainwater throughout testing had a SC value of 1.86 mS/cm, and the effluent from the column leached first with simulated rainwater at pH 2.6 and then with ambient simulated rainwater had a SC measurement of 1.64 mS/cm. After drying for 53 days between experiments, the conductivities had increased to 5.21 and 7.53 mS/cm, respectively. With the exception of Material A (carbonaceous), all of the materials originally leached with simulated rainwater at pH 2.6 had a higher SC at the start of the column leaching tests with drying than the same sample originally leached with ambient simulated rainwater. After the first leach cycle with ambient simulated rainwater, the conductivities of columns containing Materials A, B and D became nearly equal.



**Figure 14** — Specific conductivity of leach effluents from columns containing Material F.

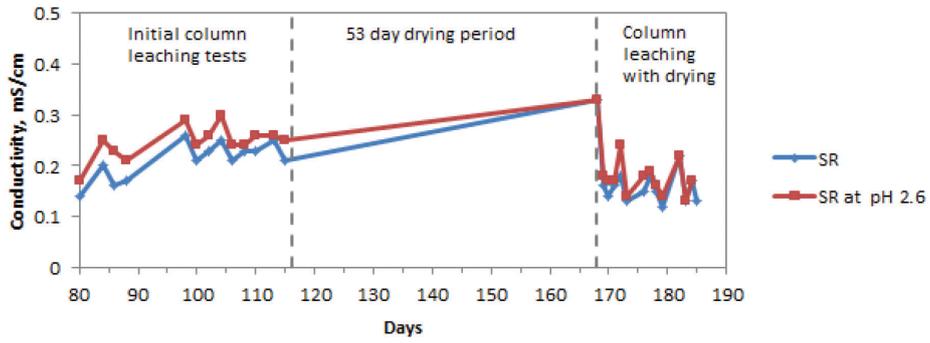


Figure 15 – SC of Material A effluent over entire leaching period.

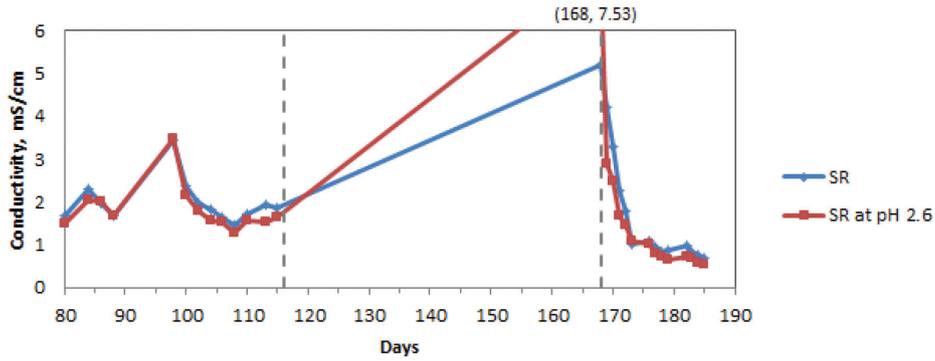


Figure 16 – SC of Material B effluent over entire leaching period. Points outside of the y-axis range are labeled at the top of the graph.

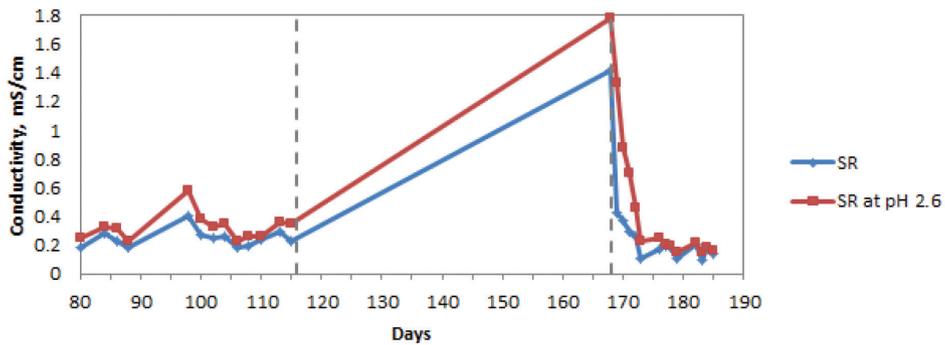


Figure 17 – SC of Material C effluent over entire leaching period.

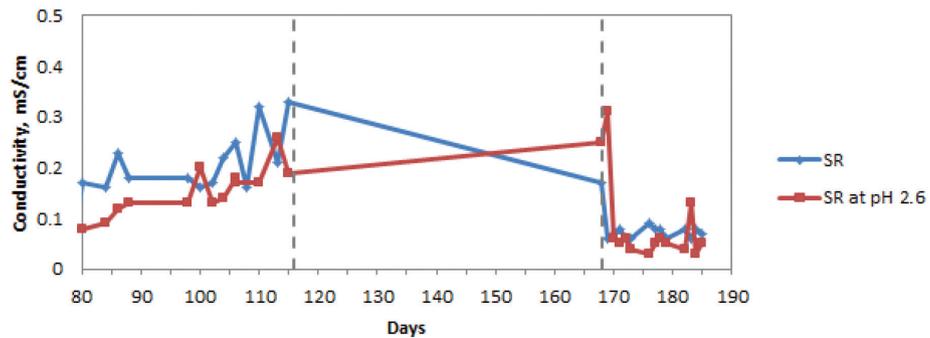


Figure 18 – SC of Material D effluent over entire leaching period.

Approximately five leach cycles were required before the columns containing Material C had nearly equal SC readings. Up until the fifth leach cycle of the drying experiments (day 173), the effluent from the column originally leached with simulated rainwater at pH 2.6 was consistently higher than the effluent from the column leached with ambient simulated rainwater. It is also interesting to note that these columns consist of layers of Materials A and B. However, during these modified leaching experiments, the trends viewed in Material C (Fig. 17) very closely resemble the trends seen in Material B (Fig. 16), which varies from the initial column leaching tests when Material C behaved more like Material A (Figs. 10 and 11).

## Discussion

The generation of SC, and thus TDS, is an intrinsic characteristic of spoil piles. Though spoil materials from Central Appalachian coal mines were studied, the problem of SC generation spans across the mining industry. The results of this study show that it is possible to reduce the SC generating potential of some materials by accelerated leaching. Specifically, shale (Material F in Fig. 14) and composite gray sandstone (Material D in Fig. 12) tested here responded favorably to the treatment.

Practically, both the material properties and leach solution characteristics would be important in determining the effectiveness of an accelerated leaching program. This point is exemplified in the present work by the weathered sandstone (Material E in Fig. 13). During the initial column leaching tests, when ambient simulated rainwater was used as the leach solution, no SC was leached from the sample. However, when using simulated rainwater at pH 2.6 and alternating the leach solutions, the SC of the effluent continued to increase. Once the leach solution for these two columns was changed to ambient simulated rainwater, the SC of the leach effluent quickly decreased. Based on the sample's reaction with ambient simulated rainwater, applying an acidic leach solution to the sample did not show any benefits compared with leaving the material in the simulated traditional spoil pile. As the sample was subjected to prior weathering, it is likely that many of the dissolvable ions had already been removed so that leaching with an acidic solution was not necessary.

For all of the materials, leaching with the acidic solution prior to leaching with ambient simulated rainwater did not cause the SC after the change to be any higher than the columns consistently leached with ambient simulated rainwater. This observation was very clear in the columns containing Materials A and B (Figs. 5 and 6, respectively). Though the SC of the effluents of the samples leached with simulated rainwater at pH 2.6 or alternating solutions tended to be higher than samples leached with ambient simulated rainwater, after all columns were switched to the ambient simulated rainwater solution, the conductivities of all three columns converged and were equivalent for the remainder of testing. Therefore, though leaching with simulated rainwater at pH 2.6 or the alternating solutions did not help accelerate the release of ions for these samples, it did not cause a continued release of ions after the leaching solution was changed to ambient simulated rainwater. Additionally, after the change to the ambient simulated rainwater leach solution, there were no (measurable) adverse effects to the samples originally leached with acidic solutions compared with the columns leached with ambient simulated rainwater throughout the study.

The most significant impact on SC seemed to be related

to whether or not the samples remained wet between leach cycles. Material B (iron-rich composite) was especially sensitive to drying, as seen in the sharp increase in SC after the 25-day drying period (Fig. 6). Samples leached with acidic solutions tended to be slightly more sensitive to drying and showed larger increases in SC upon re-wetting than columns leached with ambient simulated rainwater. Due to the design of the columns, only a small surface area was exposed at the top and bottom of each column. Therefore, the samples did not have time to fully dry when leaching frequency was once per day or every other day (even considering that leaching was typically not conducted during weekends). During the 25-day period where no leaching occurred, the samples were completely dry. Compared with the starting SC values, SC measurements in all the columns had greatly decreased just before the 25-day drying period. After drying, increases in SC were measured for all materials, with the exception of Material E (weathered sandstone). This drying period caused many of the samples to reactivate, as seen by the increasing trends in SC in all of the columns containing Material A (carbonaceous), the columns containing Material D (composite gray sandstone) leached with ambient simulated rainwater and simulated rainwater at pH 2.6, and the column containing Material F (shale) leached with ambient simulated rainwater, observed once leaching resumed on day 77.

Though increases in SC were seen in these "reactivated" materials until the end of the initial column leach tests, the same trends were not seen after the 53-day drying period, which occurred between the initial tests and the leaching tests with drying. The leaching with drying tests attempted to simulate the effects of material drying that might be associated with certain natural or operational conditions in the field. While no major increases or decreases in SC values were observed after the initial washing out of dissolved solids during the leaching tests with drying, it is noteworthy that the final SC values at the end of the leaching experiments with drying were lower than the SC values at the end of the initial column leaching tests. This overall decrease could be due to continued leaching of the columns, the extended drying time between the initial and leach-with-drying tests helping to stabilize the materials, or inadequate drying between cycles, leaving moisture in the columns. By keeping a certain moisture level within the columns, low SC could be maintained. Leaching the columns multiple times per day essentially "washed out" any dissolvable solids, resulting in low SC values. The low SC values could then be maintained by leaching the columns once per day, as long as the material did not dry out between leach cycles. In a real-world setting, extended dry periods could produce spikes of SC during rainstorms or leach cycles, which might have an impact on water quality or ecosystems downstream.

## Conclusions

This work investigated whether controlled leaching could be used to accelerate weathering of mine spoil materials common to Central Appalachia. Based on the results presented here, it is indicated that for some spoil materials it is possible to use acidic solutions to enhance the release of ions for short periods of consistent leaching, and that after the leaching period, the material is relatively more stable in regards to SC generation than before leaching or if it had been leached under natural conditions, that is, by rainwater. Moreover, the results also indicate that fully drying some materials may cause the spoil to activate and thus increase

SC and TDS generation upon re-wetting.

Further work is still needed to identify what types of materials are most favorable for accelerated leaching treatment and to characterize how field conditions might affect the long-term chemical stability of such materials. It is also recommended that columns with a surface area to depth ratio similar to traditional spoil piles be studied to better understand how the materials would dry in simulated field conditions. These tests could also help confirm that by keeping the material damp, leaching activity is decreased. In such a case, it may be beneficial for industry controlled fill moisture levels, at least for some period of time, to reduce TDS generation. Additionally, significant scale-up work would be needed to determine the feasibility of this sort of treatment program on a full scale, including investigating the effects of fill design, leach solution chemistry and recycling, and effluent treatment and disposal.

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